Hydrogen/Oxygen Reactions at High Pressures and Intermediate Temperatures: Flow Reactor Experiments and Kinetic Modeling

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Hydrogen/Oxygen Reactions at High Pressures and Intermediate Temperatures: Flow Reactor Experiments and Kinetic Modeling

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Abstract

A series of experimental and numerical investigations into hydrogen oxidation at high pressures and intermediate temperatures has been conducted. The experiments were carried out in a high pressure laminar flow reactor at 50 bar pressure and a temperature range of 600–900 K. The equivalence ratio of the mixture was varied from oxidizing to reducing conditions. Moreover, a series of experiments in an oxygen atmosphere instead of a nitrogen atmosphere has been done. A reaction mechanism based on a recent work by Burke et al. has been developed. In addition to modeling of the present experiments, the mechanism is used to simulate published data on ignition delay time and laminar burning velocity of hydrogen. The flow reactor results show that at reducing, stoichiometric, and oxidizing conditions, conversion starts at temperatures of 750–775 K, 800–825 K, and 800–825 K, respectively. In oxygen atmosphere, ignition occurs at the temperature of 775–800 K. In general, the present model provides a good agreement with the measurements in the flow reactor and with recent data on laminar burning velocity and ignition delay time.

Experimental Setup – Laminar Flow Reactor

- Quartz reactor to minimize surface reactions
- Steel pressure shell to achieve high pressures
- Temperature: 600–900 K
- Pressure: 50 bar
- Flow: 3.06 NL/min
- Isothermal Zone Length: 42–44 cm
- Residence time: 6.3–8.0 s
- Measurement via a GC and a Gas Analyzer

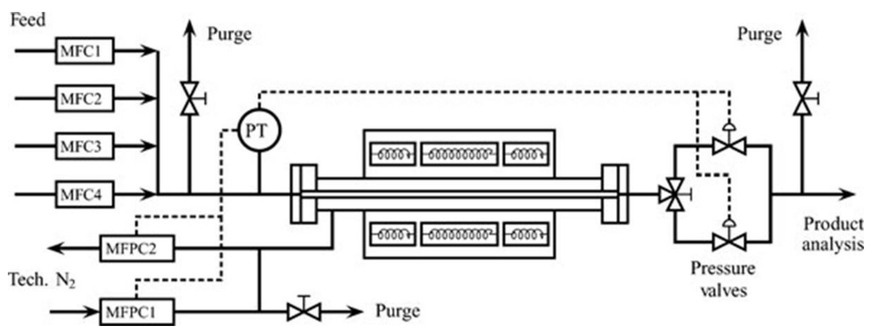

Fig 1. Schematic diagram of the high pressure laminar flow reactor

Reaction Kinetics Model

- Developed based on the mechanism by Burke et al. [1]
- Updated rates for reactions of:

  $OH + OH = O + H_2O$

  $HO_2 + OH = H_2O + O_2$

  $HO_2 + HO_2 = H_2O_2 + O_2$
- Solution via *Chemkin-Pro*

Results (The Flow Reactor)

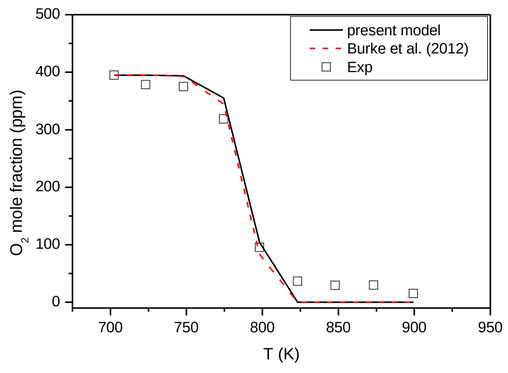

Fig 2. Results of reducing experiments (0.95% H_2 and 0.04% O_2 in N_2, Φ=12.07) at 50 bar pressure.

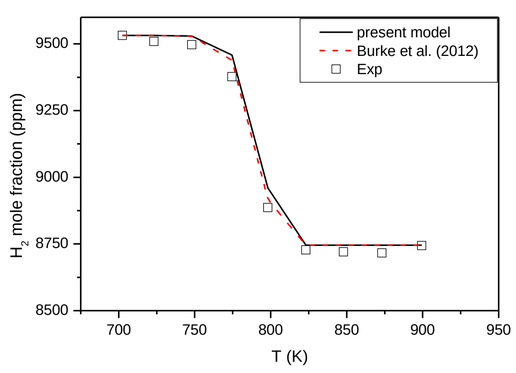

Fig 3. Results of reducing experiments (0.95% H_2 and 0.04% O_2 in N_2, Φ=12.07) at 50 bar pressure.

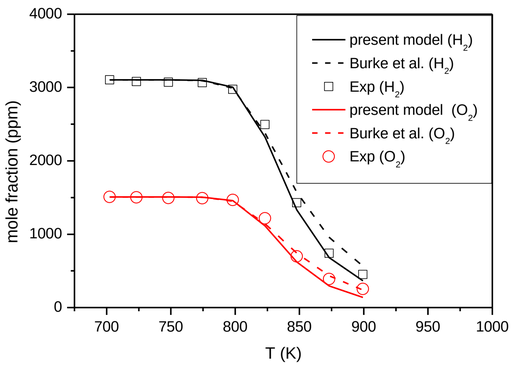

Fig 4. Results of stoichiometric experiments (0.31% H_2 and 0.15% O_2 in N_2, Φ=1.03) at 50 bar pressure.

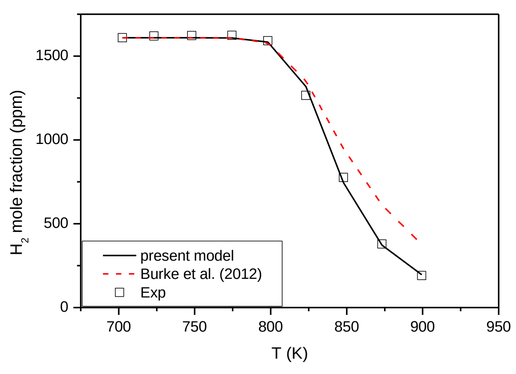

Fig 5. Results of oxidizing experiments (0.16% H_2 and 1.60% O_2 in N_2, Φ=0.05) at 50 bar pressure.

Results (The Flow Reactor)

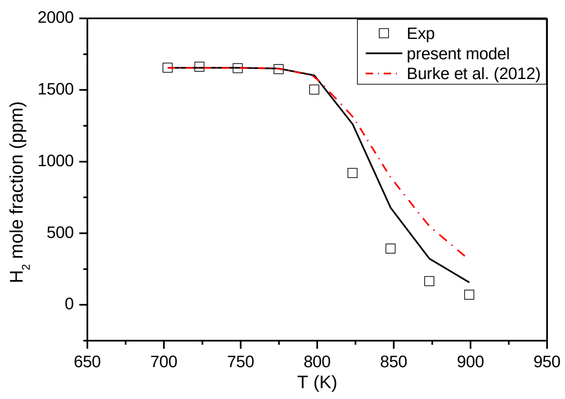

Fig 6. Results of experiments in oxygen atmosphere (0.17% H_2 and 5.91% N_2 in O_2) at 50 bar pressure.

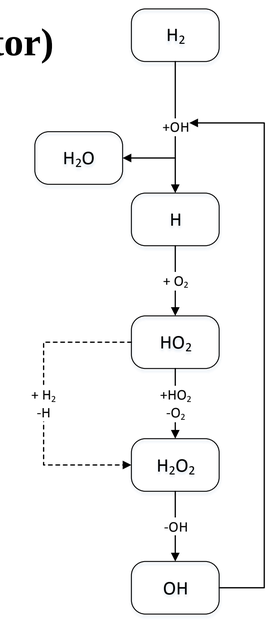

Fig 7. Major consumption path of hydrogen.

Results (Comparison to Available Data)

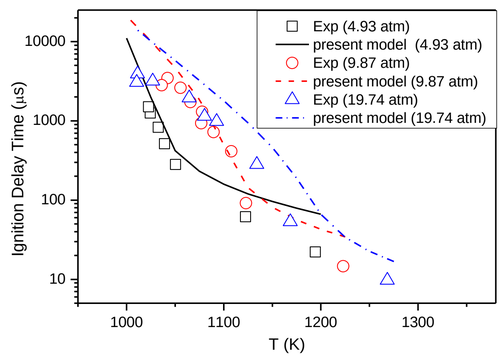

Fig 8. Ignition delay time of $H_2/O_2/Ar$ at Φ= 0.5 (X_{Argon}=93.1%). Experimental results from [2].

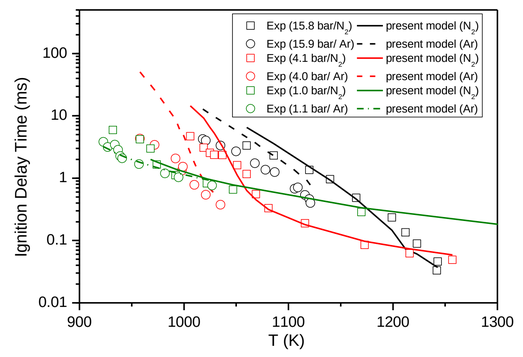

Fig 9. Ignition delay time of $H_2/O_2/Ar$ and $H_2/O_2/N_2$ at Φ= 0.5. Experimental results for Ar from [3] and for N_2 from [4].

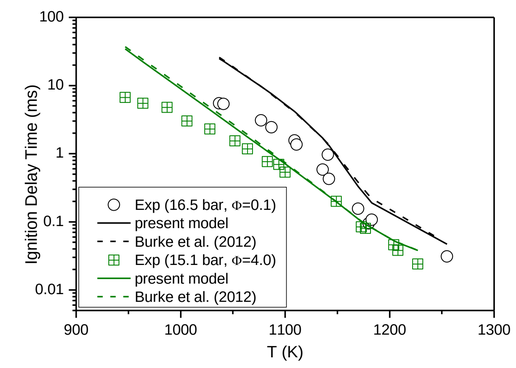

Fig 10. Ignition delay time of $H_2/O_2/Ar$ at Φ= 1.0 and 4.0. Experimental results from [4].

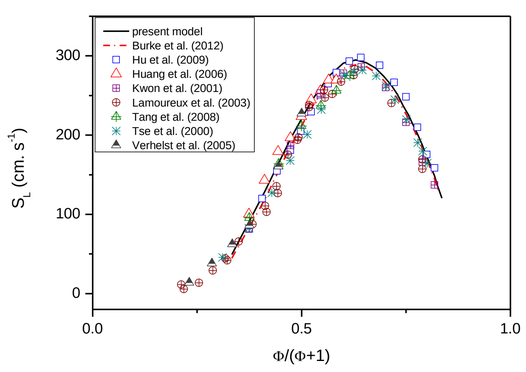

Fig 11. Unstretched laminar burning velocity of H_2/Air at initial conditions of 298 K and 1 atm. Lines denote model predictions and symbols mark experimental data from literatures.

Summary

The flow reactor results show that at reducing, stoichiometric, and oxidizing conditions, conversion starts at temperatures above 750 K, 800 K, and 800 K, respectively. In oxygen atmosphere, ignition occurs at temperatures above 775 K. The changes in the model improve its prediction especially at oxidizing conditions. Ignition delay time of hydrogen shows a non-linear trend versus pressure especially at low temperatures. The present chemical scheme used in a constant u & v model is able to predict the trend reasonably well while for a more accurate prediction at low temperatures, it is required to consider device-dependent pressure (and temperature) rise before the ignition. Predictions of laminar burning velocity by the model are within the uncertainty of the experiments. In general, the present model provides a better agreement to the measurements comparing to the base model.

References

[1] M. P. Burke, M. Chaos, Y. Ju, F. L. Dryer, S. J. Klippenstein, Int. J. Chem. Kinet. 44 (2012) 444–474.
[2] Y. Zhang, Z. Huang, L. Wei, J. Zhang, C. K. Law, Combust. Flame 159 (2012) 918–931.
[3] J. Herzler, C. Naumann, Proc. Combust. Inst. 32 (2009) 213–220.
[4] A. Keromnes, W. K. Metcalfe, K. A. Heufer, N. Donohoe, A. K. Das, C.-J. Sung, J. Herzler, C. Naumann, P. Griebel, O. Mathieu, M. C. Krejci, E. L. Petersen, W. J. Pitz, H. J. Curran, Combust. Flame 160 (2013) 995 – 1011.